

PCT

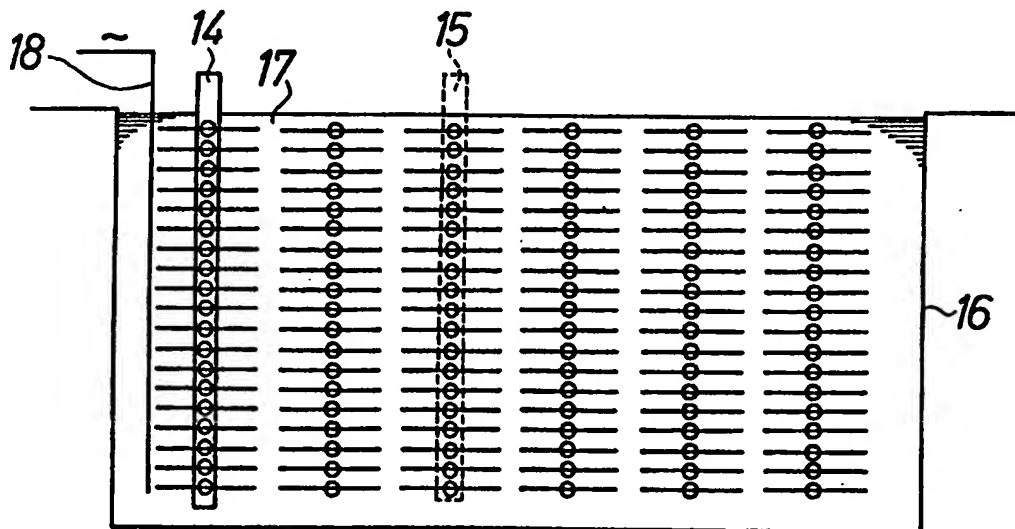
WORLD INTELLECTUAL PROPERTY ORGANIZATION  
International Bureau



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification <sup>6</sup> : <b>C01B 13/11</b>	<b>A1</b>	(11) International Publication Number: <b>WO 97/01507</b> (43) International Publication Date: 16 January 1997 (16.01.97)
(21) International Application Number: PCT/SE96/00839 (22) International Filing Date: 26 June 1996 (26.06.96) (30) Priority Data: 9502339-6 28 June 1995 (28.06.95) SE (71)(72) Applicant and Inventor: ARLEMARK, Jan [SE/SE]; Vikensväg 73 B, S-236 41 Höllviken (SE). (74) Agents: STRÖM, Tore et al.; Ström & Gulliksson AB, P.O. Box 4188, S-203 13 Malmö (SE).		(81) Designated States: AL, AM, AT, AU, AZ, BB, BG, BR, BY, CA, CH, CN, CZ, DE, DK, EE, ES, FI, GB, GE, HU, IS, JP, KE, KG, KP, KR, KZ, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, TJ, TM, TR, TT, UA, UG, US, UZ, VN, ARIPO patent (KE, LS, MW, SD, SZ, UG), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).  Published With international search report. With amended claims.

(54) Title: A METHOD AND DEVICE FOR PRODUCING OZONE



(57) Abstract

Ozone is produced in that oxygen is subjected to a high frequency alternating current with high voltage over a dielectricum. Oxygen is admitted to a narrow space between two plates (2) of aluminum oxide, and the current is applied to an electrode net (3) in the space, whereas the opposite sides of the plates are earthed and cooled.

**FOR THE PURPOSES OF INFORMATION ONLY**

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AM	Armenia	GB	United Kingdom	MW	Malawi
AT	Austria	GE	Georgia	MX	Mexico
AU	Australia	GN	Guinea	NE	Niger
BB	Barbados	GR	Greece	NL	Netherlands
BE	Belgium	HU	Hungary	NO	Norway
BF	Burkina Faso	IE	Ireland	NZ	New Zealand
BG	Bulgaria	IT	Italy	PL	Poland
BJ	Benin	JP	Japan	PT	Portugal
BR	Brazil	KE	Kenya	RO	Romania
BY	Belarus	KG	Kyrgyzstan	RU	Russian Federation
CA	Canada	KP	Democratic People's Republic of Korea	SD	Sudan
CF	Central African Republic	KR	Republic of Korea	SE	Sweden
CG	Congo	KZ	Kazakhstan	SG	Singapore
CH	Switzerland	LI	Liechtenstein	SI	Slovenia
CI	Côte d'Ivoire	LK	Sri Lanka	SK	Slovakia
CM	Cameroon	LR	Liberia	SN	Senegal
CN	China	LT	Lithuania	SZ	Swaziland
CS	Czechoslovakia	LU	Luxembourg	TD	Chad
CZ	Czech Republic	LV	Latvia	TG	Togo
DE	Germany	MC	Monaco	TJ	Tajikistan
DK	Denmark	MD	Republic of Moldova	TT	Trinidad and Tobago
EE	Estonia	MG	Madagascar	UA	Ukraine
ES	Spain	ML	Mali	UG	Uganda
FI	Finland	MN	Mongolia	US	United States of America
FR	France	MR	Mauritania	UZ	Uzbekistan
GA	Gabon			VN	Viet Nam

## A METHOD AND DEVICE FOR PRODUCING OZONE

### Technical Field

5       The present invention relates to a method and device for producing ozone by subjecting oxygene to a high frequency alternating current with high voltage over a dielectricum.

### Background of the Invention

10       Ozone gas is used to an increasing extent in many industrial applications, not the least waste water treatment, where it can destruct or remove for example complex organic molecules,  
cyanides and phenols from chemical waste,  
15       waste from paper plants and dye-mills, surfactants and detergents from washing processes, odors from waste water plants.

Waste water treated in a final step with ozone can without difficulty be reused for washing, irrigation or  
20       fire fighting.

Ozone is further used for pretreating drinking water, enhancing its quality considerably.

Last but not least ozone is used as the only acceptable alternative to chlorine for bleaching pulp in  
25       the paper making industry.

The demand for ozone is accordingly very high and steadily increasing.

It is well known that ozone is generated by so called dark or cold electrical discharges in oxygene gas or  
30       oxygene-rich gas mixtures. Presently known devices for generating ozone in satisfactory quantities and concentrations for industrial applications, however, are very voluminous and are difficult and expensive to service.

Known oxygene generators are often constructed in  
35       large, hermetically enclosed metal containers, which makes them very sensitive to moisture, both external and in the

supplied gas, from which the ozone is generated. Already at low moisture contents leak currents may occur with resultant risks for electrical short-circuits, leading to the destruction of the generator unit in question.

- 5       The objects of the invention are to increase the yield from the ozone making process, to decrease the physical dimensions of the device, to decrease the moisture sensitivity, and to minimize the stoppage times at service.

#### **The Invention**

- 10       These objects are according to the invention fulfilled in that oxygene gas is admitted to a narrow space with at least one plane wall of a material constituting the dielectricum and that the high frequency alternating current with high voltage is applied to a thread shaped  
15 electrode in the space, the opposite side of the wall being earthed and cooled.

Preferably, the narrow space has two plane walls of a material constituting dielectrica.

- 20       In one embodiment earthed metal blocks containing cooling medium are applied against the walls.

- In another embodiment an earthed cooling medium is applied directly against the walls. This is obtained in that generator units - each formed by the walls with the internal electrode - are submerged in the earthed cooling  
25 medium.

#### **Brief Description of the Drawings**

The invention will be described in further detail below reference being made to the accompanying drawings, in which

- 30       Fig 1 is a schematic illustration of the basic construction of a unit in an ozone generator according to a first embodiment of the invention,

      Fig 2 is a top view of an electrode net for use in an ozone generator as illustrated in Fig 1,

Fig 3 is a side view of an ozone generator according to the first embodiment,

Fig 4 is a top view of a second embodiment of an ozone generator according to the invention,

5 Fig 5 is a side view of the embodiment according to Fig 4,

Fig 6 is a top view of a generator unit for the embodiment according to Figs 4 and 5, and

10 Fig 7 is a side view of the generator unit according to Fig 6.

#### **Detailed Description of Embodiments**

An ozone generator according to a first embodiment of the invention is schematically depicted in Figs 1-3.

As shown in Fig 1, an ozone generator unit 1  
15 comprises two plates 2 of pure aluminum oxide or alumina, a ceramic material. It is desirable to use an aluminum oxide with a purity of 99.9 % or even better. Each such plate may for example have the dimensions length 160 mm, width 115 mm and thickness 0.65 mm.

20 A metal electrode 3 in the form of a net of acidproof stainless steel or the like is arranged between the two ceramic plates 2. The net 3, also shown in Fig 2, is surrounded by a frame 3', which is made of the same material as the plates and may have a thickness of 0.5 mm  
25 providing a space with the same height between the two plates 2. The plates 2 and the frame 3' are interconnected to a gas tight unit, for example by means of glass as glue.

As an alternative to providing a separate net in the space, thread-shaped electrode material may be arranged on  
30 the inner surfaces of the plates 2.

Each generator unit 1 is provided with openings for the admission of oxygen gas ( $O_2$ ) to and the removal of formed ozone gas ( $O_3$ ) in oxygen gas from the space between the plates 2. Also, there is an electrical connection to  
35 the net 3.

In order to cool the generator unit 1 and to provide an electrical earth for the system, in which the net 3 constitutes the phase, metal blocks 4 are arranged at either side of the generator unit 1. Each metal block 4 is provided with channels 4' for a suitable cooling medium and bores 4" for the administration of gas to and from the generator unit 1. The block 4 is connected to earth and the net 3 to a source for high voltage. Neither the fluid connections nor the electrical connections are shown in the drawings.

The cooling medium circulated through the metal blocks 4 is preferably a mixture of water and glycol or another alcohol. It is circulated by means of a compressor in such amounts and speeds that the temperature rise of the medium during a passage through the block 4 can be in the magnitude of 1°C at a chosen working temperature of -5°C.

In the described system, each plate 2 functions as a dielectricum, when the net 3 is connected to an alternating current and the blocks 4 to earth. The voltage of the preferably sinusoidal alternating current may preferably be in the order of 6 000 V, but theoretically voltages as high as 25 000-30 000 V may be used, if a break-through is obtained at the level 35 000 V. The frequency of the alternating current may be in the region of 2-100 kHz, preferably 2.5 kHz.

When this current is applied to the net 3, a so called cold discharge with a corona effect is obtained through the dielectricum constituted by the plates 2 between the line-shaped net portions and the earthed surfaces of the metal blocks 4. Hereby a certain portion of the oxygen transmitted through the generator unit 1 is converted into ozone. Under the circumstances described above the result may be as high as 18-20 vol% ozone in the oxygene gas, whereas a result of only 3-11 vol% oxygene may be obtained in conventional processes. An oxygene mixture

with a higher ozone content than 18 % may be spontaneously explosive.

The oxygene may be given a certain over-pressure of for example 0.5 bar at the introduction into the generator unit 1 so as to ensure a proper transport therethrough. Also the oxygene gas may be forced to follow a winding path through the space in the generator unit 1 - by the provision of internal partition walls - so as to increase the time and distance for the oxygene in the unit.

It is preferred to keep the working temperature of the dielectricum at 20°C, although higher temperatures of maximum 60-80°C are possible, and hereby about 80 % of the supplied electric energy is transformed to heat, which has to be cooled away.

Under the described circumstances, the yield from each generator unit may be in the order of 20 g/h, which means that 50 generator units or elements are required for producing 1 kg ozone/h. This amount may for example be needed for cleaning the waste water from about 2 000 households.

Fig 3 illustrates a stack of generator units 1 and blocks 4. Such a stack may contain a plurality of units and blocks - many more than the five units and six blocks shown.

A second embodiment of an ozone generator according to the invention is shown in Figs 4-7. The main difference in relation to the first embodiment according to Figs 1-3 is that in this case the metal blocks are missing and that the generator units, in this case bearing the numeral 10, are submerged in a liquid, which both constitutes the cooling medium and the electrical earth.

A generator unit 10 for this embodiment is shown in Figs 6 and 7. The unit 10 in itself is not in principle different from the unit 1 for the first embodiment. It comprises two plates constituting dielectrica and in a

gastight space therebetween an electrode net. The unit 10 is provided with an inlet 11 for oxygene and an outlet 12 for the formed mixture of ozone and oxygene. It is also provided with an electrical connection 13 for the electrode  
5 net.

A stack of such generator units 10 may be suspended between an inlet tube 14 for oxygene and an outlet tube 15 for ozone/oxygene, the stack being submerged in a container 16 in a liquid 17. There is also an isolated electrical  
10 cable 18 for supply of the alternating current to the generator units 10. Several such stacks, for example six, each containing a relatively large number of generator units 10, may be arranged in one container 16; in the shown case the number is 19. For the sake of clarity, only one  
15 inlet tube 14 is shown with solid lines in Fig 5 and one outlet tube 15 with dotted lines. In Fig 4 the flow of oxygene to and ozone/oxygene from the stacks of generator units 10 is illustrated by means of arrows.

The liquid 17 used in the container 16, which is to  
20 be connected to earth or is to be provided with earth plates, shall be an electrically good conductor for high frequency voltages, which is the case for water. For practical reasons the same liquid can be used as was used as the cooling medium in the first embodiment. It may be of  
25 advantage to keep the liquid 17 in a certain movement in the container 16.

For a corresponding number of generator units the arrangement of the second embodiment may be more effective, i e consume less energy. It may also be volume effective  
30 in that the distance between neighbouring generator units 10 can be kept as low as 5 mm.



## CLAIMS

1. A method of producing ozone by subjecting oxygene to a high frequency alternating current with high voltage over a dielectricum, c h a r a c t e r i z e d in that  
5 oxygene gas is admitted to a narrow space with at least one plane wall (2) of a material constituting the dielectricum and that the high frequency alternating current with high voltage is applied to a thread shaped electrode (3) in the space, the opposite side of the wall being earthed and  
10 cooled.
2. A method according to claim 1, c h a r a c t e r i z e d in that the narrow space has two plane walls (2) of a material constituting dielectrica.
3. A method according to claim 2, c h a r a c t e r i z e d in that earthed metal blocks (4) containing  
15 cooling medium are applied against the walls (2).
4. A method according to claim 2, c h a r a c t e r i z e d in that an earthed cooling medium (17) is applied directly against the walls (2).
- 20 5. A device for producing ozone by subjecting oxygene to a high frequency alternating current with high voltage over a dielectricum, c h a r a c t e r i z e d by a narrow space for oxygene gas, the space having at least one plane wall (2) of a material constituting the dielectricum, by a  
25 thread shaped electrode (3) for the current in the space, and by means for earthing and cooling the opposite side of the wall.
6. A device according to claim 5, c h a r a c t e r i z e d in that an ozone generator unit (1) has two walls  
30 (2) of a ceramic material and the thread shaped electrode (3) in the space inbetween these walls.
7. A device according to claim 6, c h a r a c t e r i z e d in that the ceramic material is aluminium oxide and the electrode (3) is of acidproof stainless steel.

8. A device according to claim 6, c h a r a c t e r-  
i z e d in that an earthed metal block (4) containing  
cooling medium is arranged directly at either side of the  
generator unit (1).

5        9. A device according to claim 6, c h a r a c t e r-  
i z e d in that the generator unit (1) is submerged in an  
earthed cooling medium (17).

10       10. A device according to claim 9, c h a r a c t e r-  
i z e d in that several generator units (1) are suspended  
as a stack between an inlet tube (14) for oxygene and an  
outlet tube (15) for ozone/oxygene in the earthed cooling  
medium (17).

## AMENDED CLAIMS

[received by the International Bureau on 28 November 1996 (28.11.96);  
original claims 1-10 replaced by new claims 1-6 (1 page)]

1. A device for producing ozone by subjecting oxygene  
in a space between two walls (2) of a dielectrical material  
to a high frequency alternating current with high voltage,  
5 wherein the space contains a plane electrode (3) and the  
walls are externally earthed and cooled, c h a r a c t e r -  
i z e d in that the plane electrode (3) made of a thread  
shaped material substantially fills the narrow space for  
oxygene gas and in that the ozone generator (1) comprising  
10 the walls (2) and the electrode (3) is manufactured to a  
sealed unit.

2. A device according to claim 1, c h a r a c t e r -  
i z e d in that the two walls (2) are made of a ceramic  
material.

15 3. A device according to claim 2, c h a r a c t e r -  
i z e d in that the ceramic material is aluminium oxide  
and the electrode (3) is of acidproof stainless steel.

4. A device according to claim 2, c h a r a c t e r -  
i z e d in that an earthed metal block (4) containing  
20 cooling medium is arranged directly at either side of the  
generator unit (1).

5. A device according to claim 2, c h a r a c t e r -  
i z e d in that the generator unit (1) is submerged in an  
earthed cooling medium (17).

25 6. A device according to claim 5, c h a r a c t e r -  
i z e d in that several generator units (1) are suspended  
as a stack between an inlet tube (14) for oxygene and an  
outlet tube (15) for ozone/oxygene in the earthed cooling  
medium (17).

1/3

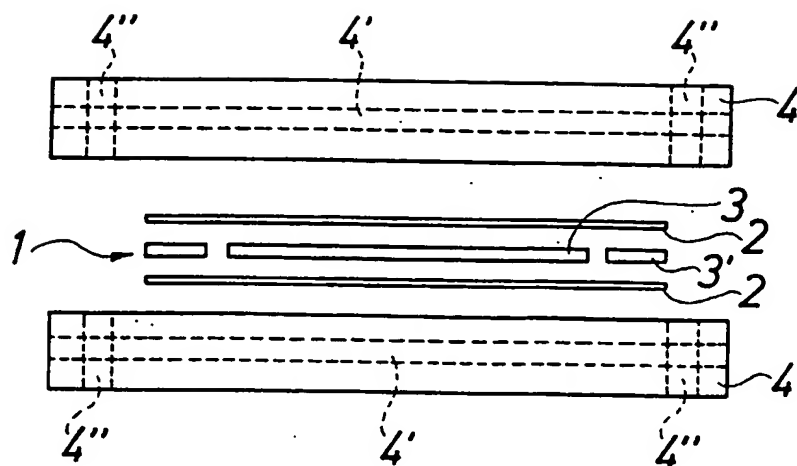


FIG. 1

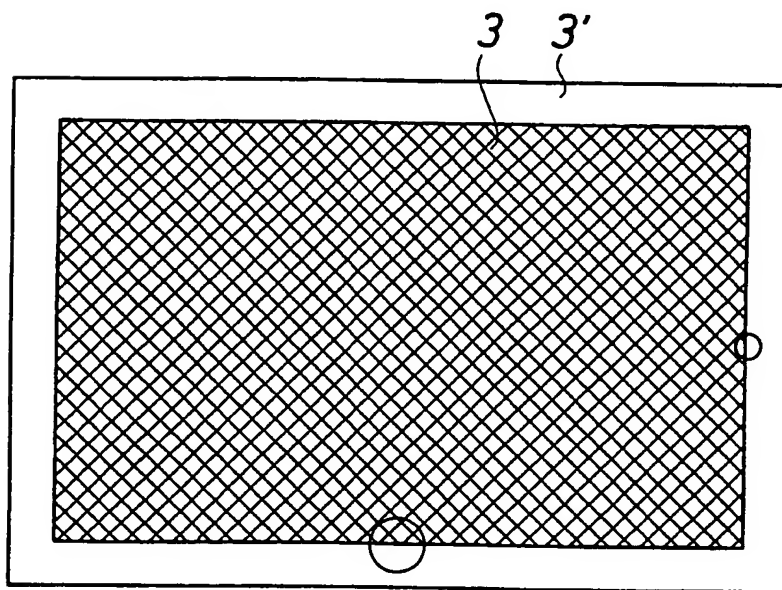


FIG. 2

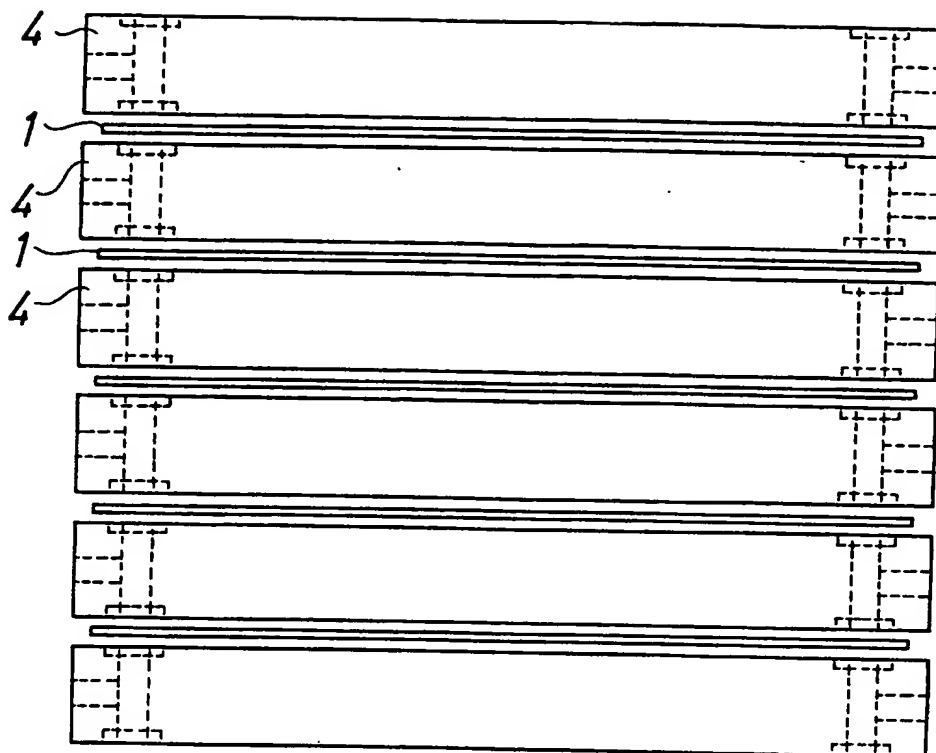


FIG. 3

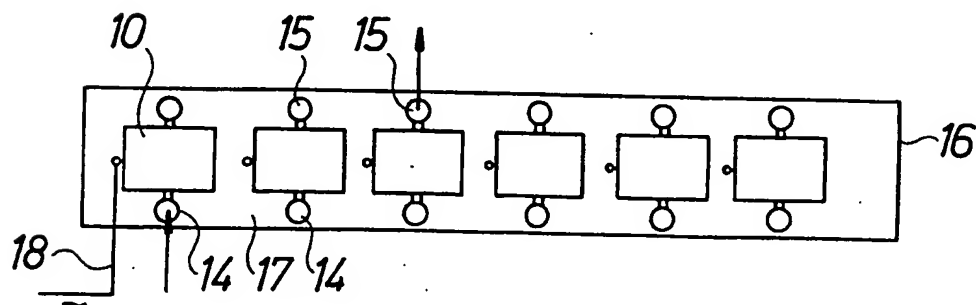


FIG. 4

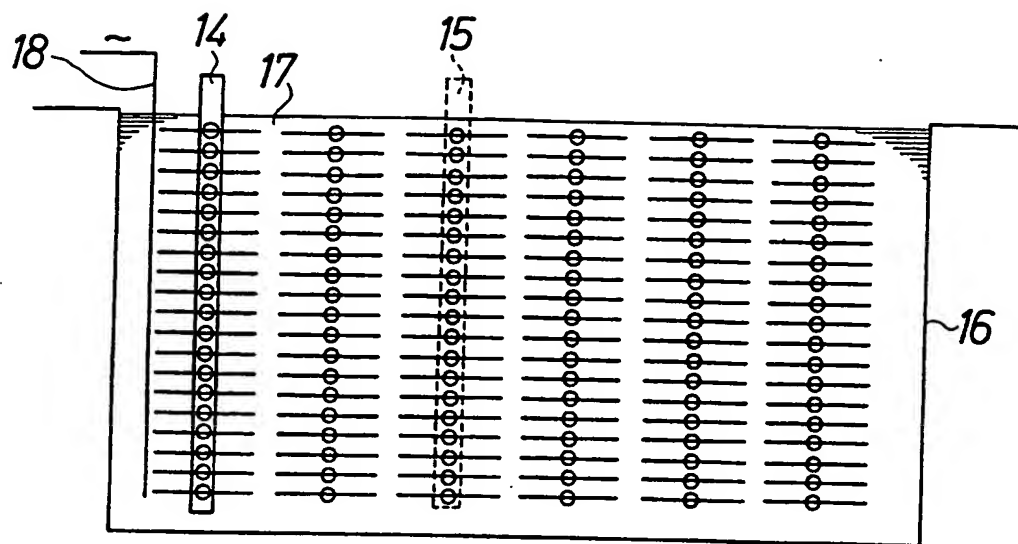


FIG. 5

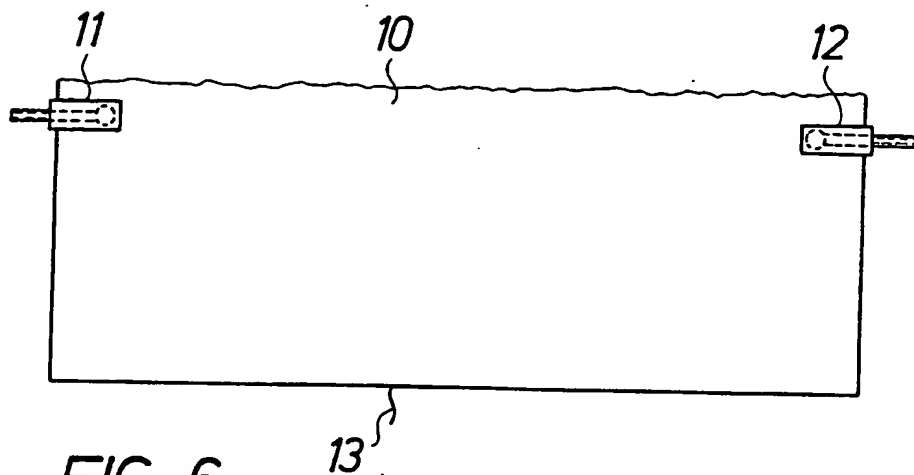


FIG. 6

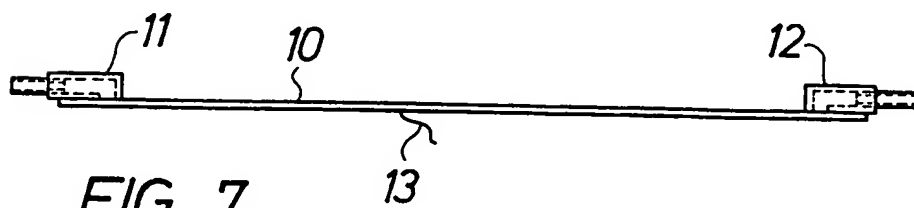


FIG. 7

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/SE 96/00839

## A. CLASSIFICATION OF SUBJECT MATTER

IPC6: C01B 13/11

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC6: C01B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

SE,DK,FI,NO classes as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

EPODOC, QUESTEL

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	CH 609309 A5 (FR. SAUTER AG), 28 February 1979 (28.02.79), page 3, column 1, line 7 - column 1, line 9; page 3, column 2, line 5 - column 2, line 7, figures 1c,3	1-3,5,6,8
Y		4,9
A		1-10
Y	CH 587187 A5 (THE ELECTRICITY COUNCIL), 29 April 1977 (29.04.77), figure 2	4,9

☐ Further documents are listed in the continuation of Box C.
 ☒ See patent family annex.

- \* Special categories of cited documents:
  - "A" document defining the general state of the art which is not considered to be of particular relevance
  - "B" earlier document but published on or after the international filing date
  - "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
  - "O" document referring to an oral disclosure, use, exhibition or other means
  - "P" document published prior to the international filing date but later than the priority date claimed
  - "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
  - "X" document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
  - "Y" document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
  - "&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
30 Sept 1996	02 -10- 1996
Name and mailing address of the ISA/ Swedish Patent Office Box 5055, S-102 42 STOCKHOLM Facsimile No. +46 8 666 02 86	Authorized officer  Johann Rigler Telephone No. +46 8 782 25 00

**INTERNATIONAL SEARCH REPORT**  
Information on patent family members

05/09/96

International application No.  
PCT/SE 96/00839

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
CH-A5- 609309	28/02/79	DE-A, C- 2541973	16/12/76
		FR-A, B- 2314139	07/01/77
		GB-A- 1495615	21/12/77
		US-A- 4034229	05/07/77
CH-A5- 587187	29/04/77	AT-B- 351491	25/07/79
		BE-A- 797256	16/07/73
		CA-A- 989349	18/05/76
		CH-A- 592023	14/10/77
		DE-A, B, C 2314398	11/10/73
		FR-A, B- 2177026	02/11/73
		FR-A, B- 2261221	12/09/75
		GB-A- 1401692	30/07/75
		JP-C- 982642	11/01/80
		JP-A- 49007187	22/01/74
		JP-B- 54016473	22/06/79
		NL-A- 7304027	25/09/73
		SE-B, C- 394657	04/07/77
		US-A- 3899685	12/08/75